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#### From the INTERNATIONAL SEARCHING AUTHORITY

To:

INTERNATIONAAL OCTROOIBUREAU B.V. Attn. DUIJVESTIJN, Adrianus J

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Applicant's or agent's file reference

**NETHERLANDS** 

MOLL

PHJP000010W0 International application No. PCT/EP 01/06294

NOTIFICATION OF TRANSMITTAL OF THE INTERNATIONAL SEARCH REPORT OR THE DECLARATION

(PCT Rule 44.1)

Date of mailing (day/month/year)	16/1	1/2001
FOR FURTHER AC	TION	See paragraphs 1 and 4 below
International filing date (day/month/year)		5/2001

Applicant

TPPI	Can				
KON	INKLIJKE PHILIPS ELECTRONICS N.V.				
1. [	The applicant is hereby notified that the International Search Report has been established and is transmitted herewith.  Filing of amendments and statement under Article 19: The applicant is entitled, if he so wishes, to amend the claims of the International Application (see Rule 46):				
	When? The time limit for filing such amendments is normally 2 months from the date of transmittal of the International Search Report; however, for more details, see the notes on the accompanying sheet.				
	Where? Directly to the International Bureau of WIPO 34, chemin des Colombettes 1211 Geneva 20, Switzerland Fascimile No.: (41–22) 740.14.35				
	For more detailed instructions, see the notes on the accompanying sheet.				
2. [	The applicant is hereby notified that no International Search Report will be established and that the declaration under Article 17(2)(a) to that effect is transmitted herewith.				
з. [	With regard to the protest against payment of (an) additional fee(s) under Rule 40.2, the applicant is notified that:  the protest together with the decision thereon has been transmitted to the International Bureau together with the applicant's request to forward the texts of both the protest and the decision thereon to the designated Offices.				
	no decision has been made yet on the protest; the applicant will be notified as soon as a decision is made.				
4. F	Further action(s): The applicant is reminded of the following:				
\$	Shortly after <b>18 months</b> from the priority date, the international application will be published by the International Bureau. If the applicant wishes to avoid or postpone publication, a notice of withdrawal of the international application, or of the priority claim, must reach the International Bureau as provided in Rules 90 <i>bis</i> .1 and 90 <i>bis</i> .3, respectively, before the completion of the technical preparations for international publication.				
١	Nithin 19 months from the priority date, a demand for international preliminary examination must be filed if the applicant wishes to postpone the entry into the national phase until 30 months from the priority date (in some Offices even later).				
١	Vithin 20 months from the priority date, the applicant must perform the prescribed acts for entry into the national phase before all designated Offices which have not been elected in the demand or in a later election within 19 months from the priority date or could not be elected because they are not bound by Chapter II.				

Name and mailing address of the International Searching Authority

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Authorized officer

Toñi Muñoz-Manneken

Form PCT/ISA/220 (July 1998)

#### NOTES TO FORM PCT/ISA/220

These Notes are intended to give the basic instructions concerning the filing of amendments under article 19. The Notes are based on the requirements of the Patent Cooperation Treaty, the Regulations and the Administrative Instructions under that Treaty. In case of discrepancy between these Notes and those requirements, the latter are applicable. For more detailed information, see also the PCT Applicant's Guide, a publication of WIPO.

In these Notes, "Article", "Rule", and "Section" refer to the provisions of the PCT, the PCT Regulations and the PCT Administrative Instructions respectively.

#### **INSTRUCTIONS CONCERNING AMENDMENTS UNDER ARTICLE 19**

The applicant has, after having received the international search report, one opportunity to amend the claims of the international application. It should however be emphasized that, since all parts of the international application (claims, description and drawings) may be amended during the international preliminary examination procedure, there is usually no need to file amendments of the claims under Article 19 except where, e.g. the applicant wants the latter to be published for the purposes of provisional protection or has another reason for amending the claims before international pbulication. Furthermore, it should be emphasized that provisional protection is available in some States only.

#### What parts of the international application may be amended?

Under Article 19, only the claims may be amended.

During the international phase, the claims may also be amended (or further amended) under Article 34 before the International Preliminary Examining Authority. The description and drawings may only be amended under Article 34 before the International Examining Authority.

Upon entry into the national phase, all parts of the international application may be amended under Article 28 or, where applicable, Article 41.

#### When?

Within 2 months from the date of transmittal of the international search report or 16 months from the priority date, whichever time limit expires later. It should be noted, however, that the amendments will be considered as having been received on time if they are received by the International Bureau after the expiration of the applicable time limit but before the completion of the technical preparations for international publication (Rule 46.1).

#### Where not to file the amendments?

The amendments may only be filed with the International Bureau and not with the receiving Office or the International Searching Authority (Rule 46.2).

Where a demand for international preliminary examination has been its filed, see below.

#### How?

Either by cancelling one or more entire claims, by adding one or more new claims or by amending the text of one or more of the claims as filed.

A replacement sheet must be submitted for each sheet of the claims which, on account of an amendment or amendments, differs from the sheet originally filed.

All the claims appearing on a replacement sheet must be numbered in Arabic numerals. Where a claim is cancelled, no renumbering of the other claims is required. In all cases where claims are renumbered, they must be renumbered consecutively (Administrative Instructions, Section 205(b)).

The amendments must be made in the language in which the international application is to be published.

#### What documents must/may accompany the amendments?

Letter (Section 205(b)):

The amendments must be submitted with a letter.

The letter will not be published with the international application and the amended claims. It should not be confused with the "Statement under Article 19(1)" (see below, under "Statement under Article 19(1)").

The letter must be in English or French, at the choice of the applicant. However, if the language of the international application is English, the letter must be in English; if the language of the international application is French, the letter must be in French.

Notes to Form PCT/ISA/220 (first sheet) (January 1994)

#### NOTES TO FORM PCT/ISA/220 (ontinued)

The letter must indicate the differences between the claims as filed and the claims as amended. It must, in particular, indicate, in connection with each claim appearing in the international application (it being understood that identical indications concerning several claims may be grouped), whether

- (i) the claim is unchanged;
- (ii) the claim is cancelled;
- (iii) the claim is new;
- (iv) the claim replaces one or more claims as filed;
- (v) the claim is the result of the division of a claim as filed.

### The following examples illustrate the manner in which amendments must be explained in the accompanying letter:

- [Where originally there were 48 claims and after amendment of some claims there are 51]:
   "Claims 1 to 29, 31, 32, 34, 35, 37 to 48 replaced by amended claims bearing the same numbers; claims 30, 33 and 36 unchanged; new claims 49 to 51 added."
- [Where originally there were 15 claims and after amendment of all claims there are 11]:
   "Claims 1 to 15 replaced by amended claims 1 to 11."
- [Where originally there were 14 claims and the amendments consist in cancelling some claims and in adding new claims]:
   "Claims 1 to 6 and 14 unchanged; claims 7 to 13 cancelled; new claims 15, 16 and 17 added." or
   "Claims 7 to 13 cancelled; new claims 15, 16 and 17 added; all other claims unchanged."
- 4. [Where various kinds of amendments are made]: "Claims 1-10 unchanged; claims 11 to 13, 18 and 19 cancelled; claims 14, 15 and 16 replaced by amended claim 14; claim 17 subdivided into amended claims 15, 16 and 17; new claims 20 and 21 added."

#### "Statement under article 19(1)" (Rule 46.4)

The amendments may be accompanied by a statement explaining the amendments and indicating any impact that such amendments might have on the description and the drawings (which cannot be amended under Article 19(1)).

The statement will be published with the international application and the amended claims.

#### It must be in the language in which the international appplication is to be published.

It must be brief, not exceeding 500 words if in English or if translated into English.

It should not be confused with and does not replace the letter indicating the differences between the claims as filed and as amended. It must be filed on a separate sheet and must be identified as such by a heading, preferably by using the words "Statement under Article 19(1)."

It may not contain any disparaging comments on the international search report or the relevance of citations contained in that report. Reference to citations, relevant to a given claim, contained in the international search report may be made only in connection with an amendment of that claim.

#### Consequence if a demand for international preliminary examination has already been filed

If, at the time of filing any amendments under Article 19, a demand for international preliminary examination has already been submitted, the applicant must preferably, at the same time of filing the amendments with the International Bureau, also file a copy of such amendments with the International Preliminary Examining Authority (see Rule 62.2(a), first sentence).

#### Consequence with regard to translation of the international application for entry into the national phase

The applicant's attention is drawn to the fact that, where upon entry into the national phase, a translation of the claims as amended under Article 19 may have to be furnished to the designated/elected Offices, instead of, or in addition to, the translation of the claims as filed.

For further details on the requirements of each designated/elected Office, see Volume II of the PCT Applicant's Guide.



# **PCT**

### **INTERNATIONAL SEARCH REPORT**

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference PHJP000010W0		of Transmittal of International Search Report 220) as well as, where applicable, item 5 below.
International application No.	International filing date (day/month/year)	(Earliest) Priority Date (day/month/year)
PCT/EP 01/06294	31/05/2001	31/05/2000
Applicant		
KONINKLIJKE PHILIPS ELECT	RONICS N.V.	
This International Search Report has bee according to Article 18. A copy is being to	n prepared by this International Searching Au ansmitted to the International Bureau.	thority and is transmitted to the applicant
This International Search Report consists  [X] It is also accompanied by	of a total of sheets. a copy of each prior art document cited in thi	s report.
Basis of the report		
	international search was carried out on the balless otherwise indicated under this item.	asis of the international application in the
the international search w Authority (Rule 23.1(b)).	vas carried out on the basis of a translation of	the international application furnished to this
b. With regard to any <b>nucleotide ar</b> was carried out on the basis of th		international application, the international search
=	onal application in written form.	
	ernational application in computer readable for	rm.
	o this Authority in written form.	
the statement that the su	o this Authority in computer readble form.  bsequently furnished written sequence listing	does not go beyond the disclosure in the
1 — "	as filed has been furnished. ormation recorded in computer readable form	is identical to the written sequence listing has been
2. Certain claims were fou	ınd unsearchable (See Box I).	
3. Unity of invention is lac	sking (see Box II).	
4. With regard to the <b>title</b> ,	,	
X the text is approved as s	ubmitted by the applicant.	
the text has been established	shed by this Authority to read as follows:	
5. With regard to the <b>abstract</b> ,  The text is approved as s	ubmitted by the applicant.	
	shed, according to Rule 38.2(b), by this Authored date of mailing of this International search re	ority as it appears in Box III. The applicant may, eport, submit comments to this Authority.
6. The figure of the drawings to be pub	olished with the abstract is Figure No.	
as suggested by the app	licant.	X None of the figures.
because the applicant fa	iled to suggest a figure.	
because this figure bette	r characterizes the invention.	

#### (12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

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For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.



PCT/EP01/06294

#### A dielectric ceramic composite

#### Technical Field

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The invention relates to a dielectric ceramic composite containing (BaNdSm)TiO<sub>3</sub>, and to an electronic device.

#### 5 Background of the Invention

In recent years, in the field of the high frequency equipments such as mobile phones, their size has become smaller, their performance has become higher and their price has become lower. According to such tendency, a smaller size, high-performance and less expensive price are also desired for dielectric resonators that are used in such high frequency equipments. In particular, a high relative dielectric constant and a high Q factor are required for dielectric ceramic composites that are used as materials for those dielectric resonators. A dielectric ceramic composite that meets such requirement is disclosed in Japan Patent Application Laid-Open No. 1995-104949. If dielectric ceramic composites in accordance with the above-referenced patent application are used to produce, for example, multi-layer ceramic capacitors, it is possible to obtain capacitors having good characteristics.

As materials of internal electrodes formed in the multi-layer ceramic capacitors, precious metals such as Pd, Pt and Au are used. However, if those precious metals are used as materials for the internal electrodes, there exists a problem that the material cost may become expensive. Accordingly, the use of less expensive metals such Ag may be considered instead of the use of the precious metals. However, the melting point of the Ag is about 960 °C whereas the sintering temperature for the dielectric ceramic composite disclosed in the above-referenced patent application is close to 1400 °C. So, if a multi-layer ceramic capacitor is produced by combination of the Ag with the dielectric ceramic composite disclosed in the above-referenced patent application, there is a problem that the Ag may be melted out during the sintering process of the dielectric ceramic composite.

In view of the above-described background, it is an objective of the invention to provide a dielectric ceramic composite that can be sintered at a low temperature.

#### Summary of the Invention

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In order to achieve the above-described objective, the dielectric ceramic composite according to the present invention is characterised by comprising (BaNdSm)TiO<sub>3</sub>, ZnO, SiO<sub>2</sub>, CuO, Al<sub>2</sub>O<sub>3</sub>, MgO, B<sub>2</sub>O<sub>3</sub>, Bi<sub>2</sub>O<sub>3</sub> and either BaCO<sub>3</sub> or BaO. By including these materials, it becomes possible to sinter the dielectric ceramic composite at a low temperature.

In the dielectric ceramic composite according to the present invention, the total weight of the said ZnO, SiO<sub>2</sub>, CuO, Al<sub>2</sub>O<sub>3</sub>, MgO, B<sub>2</sub>O<sub>3</sub>, Bi<sub>2</sub>O<sub>3</sub> and either BaCO<sub>3</sub> or BaO is preferably about 20% through 30% of the weight of the said (BaNdSm)TiO<sub>3</sub>. Additionally, a ratio of the total weight of the said ZnO, SiO<sub>2</sub>, CuO, Al<sub>2</sub>O<sub>3</sub>, MgO, B<sub>2</sub>O<sub>3</sub> and either BaCO<sub>3</sub> or BaO with the weight of the said Bi<sub>2</sub>O<sub>3</sub> is preferably in a range of 0.67 to 1.50. Such the total weight or the ratio is possible to realize the high relative dielectric constant and the high Q factor.

Furthermore, in the inventive dielectric ceramic composite, the average of the grain sizes of the said SiO<sub>2</sub>, CuO and Al<sub>2</sub>O<sub>3</sub> is preferably no more than 30 nm. With such size of the grains, it becomes possible to sinter the dielectric ceramic composite at a further lower temperature.

#### Detailed Description of the Invention

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As examples of the dielectric ceramic composite in accordance with the invention, the 1st to the 17th embodiments of the dielectric ceramic composite that are appropriate materials for single planar capacitors will be described in the following. Each of the 1st to the 17th embodiments of dielectric ceramic composites mainly contains the ceramic composite "(BaNdSm)TiO<sub>3</sub>" comprising Ba (barium), Ti (titanium), Nd (neodymium) and Sm (samarium). Furthermore, in each dielectric ceramic composite, ZnO, SiO<sub>2</sub>, CuO, Al<sub>2</sub>O<sub>3</sub>, MgO, BaO, B<sub>2</sub>O<sub>3</sub> and Bi<sub>2</sub>O<sub>3</sub> are added to the ceramic composite (BaNdSm)TiO<sub>3</sub>. The method of manufacturing the 1st to the 17th embodiments of the dielectric ceramic composites will below explained.

In manufacturing the 1st to the 17th embodiments of the dielectric ceramic composites, a ceramic composite (BaNdSm)TiO<sub>3</sub> mainly contained in the dielectric ceramic composite is first manufactured as explained below.

Firstly, BaCO<sub>3</sub>, Nd<sub>2</sub>O<sub>3</sub>, Sm<sub>2</sub>O<sub>3</sub> and TiO<sub>2</sub> are chosen as starting materials. And the each required quantity of the BaCO<sub>3</sub>, Nd<sub>2</sub>O<sub>3</sub>, Sm<sub>2</sub>O<sub>3</sub> and TiO<sub>2</sub> was weighed. In these embodiments, for BaCO<sub>3</sub>, Nd<sub>2</sub>O<sub>3</sub>, Sm<sub>2</sub>O<sub>3</sub> and TiO<sub>2</sub>, 18mo1%, 11mo1%, 4mo1% and 67mo1% are weighed respectively. After having been weighed, these weighed materials are wet-blended for three hours using zirconium beads (in the wet-blending, water is used as

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solvent) and then these blended materials are dried out. Thereafter, a mixture of BaCo3, Nd<sub>2</sub>O<sub>3</sub>, Sm<sub>2</sub>O<sub>3</sub> and TiO<sub>2</sub> obtained in such a manner are calcined for two hours at a temperature of 1,170 °C. Then, the calcined materials are wet-ground for three hours using zirconium beads (in the wet-grinding, water is used as solvent) and then the ground materials are dried out. Thus, a (BaNdSm)TiO<sub>3</sub> is manufactured.

In the next step, eight kinds of compounds of ZnO, SiO<sub>2</sub>, CuO, Al<sub>2</sub>O<sub>3</sub>, MgO, BaCO<sub>3</sub>, B<sub>2</sub>O<sub>3</sub> and Bi<sub>2</sub>O<sub>3</sub> (hereinafter, ZnO, SiO<sub>2</sub>, CuO, Al<sub>2</sub>O<sub>3</sub>, MgO, BaCO<sub>3</sub>, B<sub>2</sub>O<sub>3</sub> and Bi<sub>2</sub>O<sub>3</sub> may be simply referred to as "eight kinds of compounds A") are added to the manufactured (BaNdSm)TiO<sub>3</sub>, and then those materials are wet-blended for three hours. In the instant example, the 1st to 17th embodiments of the dielectric ceramic composites D1 to D17 are manufactured by changing the mixture rates and the grain sizes of the each compound of "the eight kinds of the compound A". Table 1 shows the compositions of the dielectric ceramic composites D1 to D12 of the 1st to 12th embodiments and Table 2 shows the compositions of the dielectric ceramic composites D13 to D17 of the 13th to 17th embodiments.

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[Table 1]

[Table 2]

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Table 1 shows the weight% of each compound added to the (BaNdSm)TiO<sub>3</sub> when the weight% of the (BaNdSm)TiO<sub>3</sub> is defined as "100". In Table 1, for the dielectric ceramic composites D1 to D5, seven kinds of compounds of ZnO, SiO<sub>2</sub>, CuO, Al<sub>2</sub>O<sub>3</sub>, MgO, BaCO<sub>3</sub> and B<sub>2</sub>O<sub>3</sub> (hereinafter, ZnO, SiO<sub>2</sub>, CuO, Al<sub>2</sub>O<sub>3</sub>, MgO, BaCO<sub>3</sub> and B<sub>2</sub>O<sub>3</sub> may be simply referred to as "seven kinds of compounds B") are added to the (BaNdSm)TiO<sub>3</sub> by 10% in weight relative to the (BaNdSm)TiO<sub>3</sub>. The remaining compound Bi<sub>2</sub>O<sub>3</sub> (hereinafter, the compound Bi<sub>2</sub>O<sub>3</sub> may be referred to as "one kind of compound C") is also added to the (BaNdSm)TiO<sub>3</sub> by 10% in weight relative to the (BaNdSm)TiO<sub>3</sub>. However, the respective weight% of each compound of "seven kinds of the compounds B" varies with each of the embodiments D1 to D5.

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Besides, in all of the dielectric ceramic composites D1 and D6 through D12 of the 1st and the 6th through 12th embodiments, the ratio of addition of "seven kinds of compounds B" is  $ZnO: SiO_2: CuO: Al_2O_3: MgO: BaCO_3: B_2O_3 = 27.9: 22.4: 5.0: 10.1: 3.0: 25.9: 5.7. However, weight%s of the "seven kinds of compounds B" as a whole vary with the dielectric ceramic composites D1 and D6 through D12, or the B/Cs representing the$ 

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weight% ratio of "seven kinds of compounds B" and "one kind of compound C" vary with the dielectric ceramic composites D1 and D6 through D12.

Table 1 also shows the average of the grain sizes for each of ZnO, SiO<sub>2</sub>, CuO, Al<sub>2</sub>O<sub>3</sub>, MgO, BaCO<sub>3</sub>, B<sub>2</sub>O<sub>3</sub> and Bi<sub>2</sub>O<sub>3</sub>. The average of the grain sizes for Bi<sub>2</sub>O<sub>3</sub> is not shown because Bi<sub>2</sub>O<sub>3</sub> dissolves into water.

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With respect to the dielectric ceramic composites D13 through D17 of the 13th through 17th embodiments shown in Table 2, the weight % of each compound equals to that of the dielectric ceramic composite D1 of the 1st embodiment but the grain size of each compound is different from that of the dielectric ceramic composite D1. Table 2 shows the grain sizes (nm) for each of the compounds used in the dielectric ceramic composites D13 through D17. Further, Table 2 also shows the grain sizes (nm) for compounds used in the dielectric ceramic composite D1.

Now, the method of manufacturing a single planar capacitor using the dielectric ceramic composites D1 through D17 will be explained. Firstly, each of the 15 dielectric ceramic composites D1 to D17 is wet-blended for three hours using zirconium beads (in the wet-blending, water is used as solvent). And next, each of blended dielectric ceramic composites D1 to D17 is dried out, and each of dried dielectric ceramic composites D1 to D17 is ground into dried powder. Then, each of the dielectric ceramic composites D1 through D17 in dried powder is granulated while adding PVA (polyvinyl alcohol) as a binder. 20 After granulating each of the dielectric ceramic composites D1 to D17, each of the granulated dielectric ceramic composites D1 to D17 is charged into the mold with the 16.5mm $\Phi$  and is molded by means of a presser by a pressure of 3 tons per square centimeter. In such way, disk-like samples having a thickness of 0.7 mm are manufactured for each of the dielectric ceramic composites D1 to D17. Thereafter, these disk-like samples are sintered for two hours in the air at the temperature of 880 to 930 °C and an Ag paste is printed on 25 each of the sintered samples and then each of the printed samples is baked at the temperature of 750 °C. Thus, single planar capacitors are manufactured.

The dielectric ceramic composites of the 1st through 17th embodiments can be sintered at the temperature of about 900 °C that is lower than the sintering temperature for the conventional dielectric ceramic composites, so that the low temperature sintering can be realized.

As described above, the dielectric ceramic composites of the 1st through 17th embodiments are used to manufacture the single planar capacitor. However, the dielectric

ceramic composite according to the present invention may be used to manufacture any other electronic devices than the single planar capacitor.

Examples of other electronic devices include ceramic multilayer capacitors, filters networks of passive components comprising planar capacitors as well as multilayer substrates, like a LTCC-substrate or a laminate. The construction of such devices is generally known per se by the person skilled in the art, various are present in the patent literature. The dielectric composite, especially in the preferred embodiment with a grain size of less than 50 nm, is very well suited for the application in substrates. First of all, it can be sintered at a temperature lower than the melting point of Cu, which is a standard electrode material in these substrates. Secondly, due to the grain size it can be mixed very well with other substituents of the substrate, such as epoxy in the case of a laminate. Thirdly, due to the high Q-factor, an electronic device with composite of the invention can be applied under high-frequency conditions, such as in telecom applications.

Moreover, although the dielectric ceramic composites of the 1st through 17th embodiments contain BaCO<sub>3</sub> in the instant example, for example BaO can be used as an alternative to BaCO<sub>3</sub>. However, since BaO is an unstable material, it may be easier to manufacture the dielectric ceramic composites by using BaCO<sub>3</sub> rather than BaO.

From now on, reference is made to Table 1 to 3 for explaining some embodiment examples of the invention. At first, embodiment examples 1 to 21 of single planar capacitors were manufactured using the dielectric ceramic composites D1 to D17 of the 1st to 17th embodiments. Table 3 shows electric characteristics of each of embodiment examples 1 to 21 of single planar capacitors.

[Table 3]

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The embodiment examples 1 to 5 in Table 3 are respective single planar capacitors that were manufactured by sintering the dielectric ceramic composites D1 of the 1st embodiment at the respective temperatures 870, 880, 900, 910 and 930 °C. The embodiment examples 6 to 16 in Table 3 are respective single planar capacitors that were manufactured by sintering the respective dielectric ceramic composites D2 to D12 of the 2nd to 12th embodiments at the temperature of 910 °C. The examples 17 to 21 in Table 3 are respective single planar capacitors that were manufactured by sintering the respective dielectric ceramic composites D13 to D17 of the 13th to 17th embodiments at the temperature of 930 °C.

Relative dielectric constants and Q factors for each of the single planar capacitors are measured using an automatic bridge-type measuring equipment in a condition of 1 MHz, 1 Vrsm. Besides, temperature dependencies of capacitance TC(ppm/degree C) shown in Table 3 are temperature dependencies at the temperature of -55 to +125 °C with reference to the capacitance at the temperature of +25 °C.

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Referring to embodiment examples 1 to 5, it is observed that capacitors having different characteristics can be manufactured by changing the sintering temperature although these examples are used the same dielectric ceramic composite D1. Now, considering a case, for example, where a capacitor is to be applied to a capacitor component of a resonator, the capacitor should preferably have such characteristics that the relative dielectric constant be no less than 70, the Q factor be no less than 2000 and the TC be within +/- 30(ppm/°C). Embodiment examples 2 to 5 of single planar capacitors have such characteristics that the relative dielectric constant is no less than 70, the Q factor is no less than 2000 and the TC is within +/- 30(ppm/°C). The sintering temperature for the embodiment examples 2 to 5 is 880 to 930 °C. Thus, it is found that capacitors appropriate for the capacitor component of the resonator could be obtained by using the temperature of 880 to 930 °C.

Next, referring to embodiment examples 4 and 6 to 9, it can be observed that all of the single planer capacitors are appropriate for the capacitor component of the resonator since all of the single planer capacitors have the characteristics that the relative dielectric constant is no less than 70, the Q factor is no less than 2000 and the TC is within +/- 30(ppm/°C). As for the dielectric ceramic composites D1 to D5 that are used for each of the single planar capacitors of the embodiment examples 4 and 6 to 9, the weight% of the "seven kinds of the compounds B" added to (BaNdSm)TiO<sub>3</sub> is all equivalent (10%) as shown in Table 1 but the ratio of each compound added to the (BaNdSm)TiO<sub>3</sub> is different in each embodiment. Therefore, for the purpose of obtaining capacitors which are appropriate for the capacitor component of the resonator, the ratio of each of compounds contained in the dielectric ceramic composite is not limited to a specific value. As a result, it can be observed that it is possible to obtain the capacitors appropriate for the capacitor component of the resonator even if the ratio of each of compounds contained in the dielectric ceramic composite is changed.

Referring to embodiment examples 4 and 10 to 12, their sintering temperatures are equally 910 °C, and further the B/Cs for the dielectric ceramic composites D1 and D6 to D8 used in capacitors of embodiment examples 4 and 10 to 12 are equally 1 (refer to Table

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1). However, weight%s of B+Cs for the dielectric ceramic composites D1 and D6 to D8 are different each other. To be more specific, the weight % of the B+C is 20% for the dielectric ceramic composite D1 used in the embodiment example 4, the weight % of the B+C is 10% for the dielectric ceramic composite D6 used in the embodiment example 10, the weight % of the B+C is 30% for the dielectric ceramic composite D7 used in the embodiment example 11, and the weight % of the B+C is 40% for the dielectric ceramic composite D8 used in the embodiment example 12. It is observed that the single planer capacitors of the embodiment examples 4 and 11 using the dielectric ceramic composites D1 and D7 (their weight ratio of B+C is within 20-30%) have the characteristics that the relative dielectric constant is no less than 70, the Q factor is no less than 2000 and the TC is within +/- 30(ppm/°C). Therefore, it is possible to manufacture capacitors that are appropriate for the capacitor component of the resonator by keeping the weight% of B+C within 20-30%.

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Referring to embodiment examples 4 and 13 to 16, although their sintering temperatures are equally 910 °C, the B/Cs of the dielectric ceramic composites D1, D9 to D12 that are used therein are different each other. To be more specific, the B/C is 1 for the dielectric ceramic composite D1 and the respective B/Cs is 1.5, 2.3, 0.67 and 0.43 for the dielectric ceramic composites D9 to D12 that are used for the embodiment examples 13-16, respectively. Among the single planar capacitors of the embodiment examples 4 and 13 to 16, the single planar capacitors of the embodiment examples 4, 13 and 15 that use the dielectric ceramic composites D1, D9 and D11 having the B/C weight % in the range of 0.67 to 1.5 show acceptable characteristics that the relative dielectric constant is no less than 70, the Q factor is no less than 2000 and the TC is within +/- 30(ppm/°C). Therefore, it is possible to manufacture capacitors that are appropriate for the capacitor component of the resonator by keeping the B/C weight % in the range of 0.67 to 1.5.

Finally, referring to embodiment examples 5 and 17 to 21, although their sintering temperatures are equally 930 °C, the grain sizes of SiO<sub>2</sub>, CuO and Al<sub>2</sub>O<sub>3</sub> of the dielectric ceramic composites D1, D13 to D17 that are used therein are different each other (see Table 2). Among the single planar capacitors of the embodiment examples 5 and 17 to 21, the single planar capacitors of the embodiment example 5 that uses the dielectric ceramic composites D1 having SiO<sub>2</sub>, CuO and Al<sub>2</sub>O<sub>3</sub> with their grain sizes within no more than 30 nm show acceptable characteristics that the relative dielectric constant is no less than 70, the Q factor is no less than 2000 and the TC is within +/- 30(ppm/°C). Therefore, it is possible to manufacture capacitors that are appropriate for the capacitor component of the resonator by keeping the grain sizes of SiO<sub>2</sub>, CuO and Al<sub>2</sub>O<sub>3</sub> within no more than 30 nm.

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As described above, the invention provides a dielectric ceramic composite that can be sitered at a low temperature.

CLAIMS:

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- 1. A dielectric ceramic composite characterised by comprising (BaNdSm)TiO<sub>3</sub>, ZnO, SiO<sub>2</sub>, CuO, Al<sub>2</sub>O<sub>3</sub>, MgO, B<sub>2</sub>O<sub>3</sub>, Bi<sub>2</sub>O<sub>3</sub> and either BaCO<sub>3</sub> or BaO.
- A dielectric ceramic composite as claimed in claim 1, characterized in that the
   total weight of the said ZnO, SiO<sub>2</sub>, CuO, Al<sub>2</sub>O<sub>3</sub>, MgO, B<sub>2</sub>O<sub>3</sub>, Bi<sub>2</sub>O<sub>3</sub> and either BaCO<sub>3</sub> or BaO is about 20% through 30% of the weight of the said (BaNdSm)TiO<sub>3</sub>.
  - 3. A dielectric ceramic composite as claimed in claim 2, characterized in that a ratio of the total weight of the said ZnO, SiO<sub>2</sub>, CuO, Al<sub>2</sub>O<sub>3</sub>, MgO, B<sub>2</sub>O<sub>3</sub> and either BaCO<sub>3</sub> or BaO with the weight of the said Bi<sub>2</sub>O<sub>3</sub> is in a range of 0.67 to 1.50.
    - 4. A dielectric ceramic composite as claimed in claim 2 or 3, characterized in that the average of the grain sizes of the said SiO<sub>2</sub>, CuO and Al<sub>2</sub>O<sub>3</sub> is no more than 30 nm.
- 15 5. An electronic device comprising the dielectric ceramic composite according to one of the claims 1-4

### INTERNATIONAL SEARCH REPORT

ernational Application No PCT/EP 01/06294

A. CLASSIFICATION OF SUBJECT MATTER
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C01G23/00

According to International Patent Classification (IPC) or to both national classification and IPC

#### B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols) IPC 7 CO4B HO1B HO1G CO1G

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, PAJ, WPI Data, CHEM ABS Data, COMPENDEX, INSPEC

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X Further documents are listed in the continuation of box C.	Patent family members are listed in annex.
Special categories of cited documents:  'A' document defining the general state of the art which is not considered to be of particular relevance  'E' earlier document but published on or after the international filing date  'L' document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)  'O' document referring to an oral disclosure, use, exhibition or other means  'P' document published prior to the international filing date but later than the priority date claimed	"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention  "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone  "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.  "&" document member of the same patent family
Date of the actual completion of the international search  2 November 2001	Date of mailing of the international search report  16/11/2001
Name and mailing address of the ISA	
European Patent Office, P.B. 5818 Patentlaan 2 NL – 2280 HV Rijswijk Tel. (+31–70) 340–2040, Tx. 31 651 epo nl, Fax: (+31–70) 340–3016	Authorized officer Raming, T



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